The effect of quenching media on the heat transfer coefficient of polycrystalline alumina

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Surface heat transfer coefficient values were measured for polycrystalline alumina quenched into water, into oil, and into liquid nitrogen. Since the measurements of the surface heat transfer coefficient h for alumina (and ceramics in general) are very limited, we compare our measurements with calculations of h for the water quench and with measurements of h on non-ceramic materials for the oil and liquid nitrogen quenches.

1. Introduction
This paper presents surface heat transfer coefficient measurements for polycrystalline alumina thermally quenched into a room-temperature water bath, a room-temperature silicone oil bath, and a liquid nitrogen bath. The heat transfer measurements utilized a parameter estimation technique and fast-response thin film thermocouples [1-4]. The measured surface heat transfer coefficients are compared with other researchers' data. The experimental results are pertinent to the calculation of thermal stresses encountered during thermal shock of ceramics [1, 2].

2. Experimental procedure
To evaluate the heat transfer coefficient of polycrystalline alumina for water quench, silicone oil quench and liquid nitrogen quench, the surface temperature changes were measured by using a thin-foil "cement-on" K-type thermocouple (CO2-K, Omega Engineering, Stamford, Connecticut). The response time of this thin-foil thermocouple is 2 to 5 ms, as determined by quenching the thermocouple itself into a room-temperature water bath. With a thin-film thermocouple attached, the specimens were heated in air in an electrical resistance furnace. The specimens were then shocked into a water bath, into a silicone oil bath, or into a liquid nitrogen bath.

The geometry of the polycrystalline alumina specimens was that of bars of rectangular cross-section, with typical dimensions of approximately 7.0 cm × 1.1 cm × 0.1 cm. The thin-film thermocouples were bonded at the centre of the 7.0 cm × 1.1 cm face of the specimens using Omega CC high-temperature cement (Omega Engineering).

The maximum continuous operating temperature of the thin-film thermocouple was 540 °C and the maximum use temperature for 10 h was 640 °C (as specified by the thermocouple vendor, Omega Engineering). The maximum furnace temperature employed during this study was 580 °C, which was maintained for time periods of less than 1 h. For the liquid nitrogen bath in particular, the maximum furnace temperature of 500 °C resulted in a maximum temperature difference of 696 °C. The transient temperature of the quenched specimens, recorded via an oscilloscope (Fig. 1), was used to calculate the surface heat transfer coefficient. Details of the experimental apparatus are given elsewhere [1, 2].

3. Estimation of heat transfer coefficient
From the transient surface temperature measurements, the surface heat transfer coefficients were estimated using a parameter estimation method [1-4]. If we assume the specimen is a thermally lumped* body, then temperature is a function of time only. A lumped body which is suddenly immersed into a quenching fluid at a temperature T_∞ is described by [1-4]

$$hA(T_\infty - T) = \rho C_p V(dT/dt) \qquad (1)$$

where h is the heat transfer coefficient, ρ is the density of the specimen, C_p is the specific heat of the specimen, t is the time, V is the volume of the specimen, A is the heated surface area of the specimen and T is the true specimen temperature.

Among the several estimation procedures available for lumped bodies [1-4], we selected a regression method which employs a polynomial form in the regression of the temperature-time data [3, 4], such that

$$\hat{T} = \beta_1 + \beta_2\left(\frac{t}{\Delta t}\right) + \cdots + \beta_p\left(\frac{t}{\Delta t}\right)^{p-1} \qquad (2)$$

where $\hat{T}$ is the estimated temperature and Δt is the time interval between readings of the thermocouple

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*Thermally lumped bodies are bodies in which the thermal conductivity is large or the characteristic length scale (volume/surface area) is small, such that the temperature of the solid is spatially uniform at any instant during the transient process. In this study, the characteristic length scale of the alumina specimens is relatively small (section 2), so we approximate the specimens as lumped bodies.
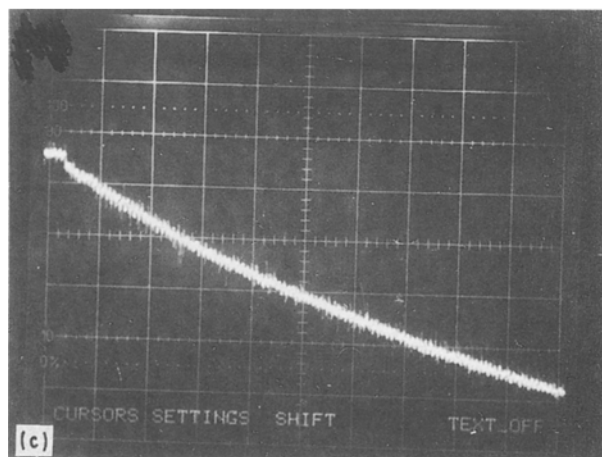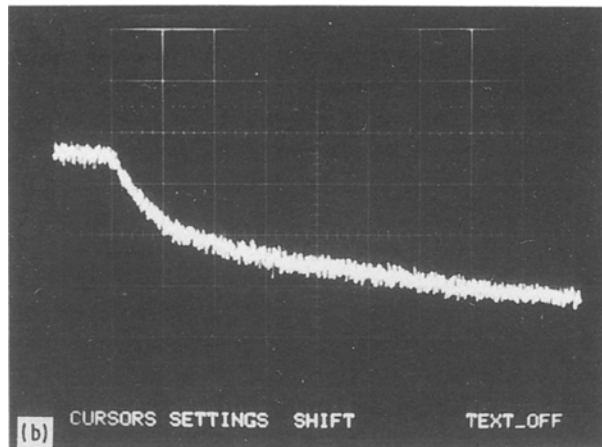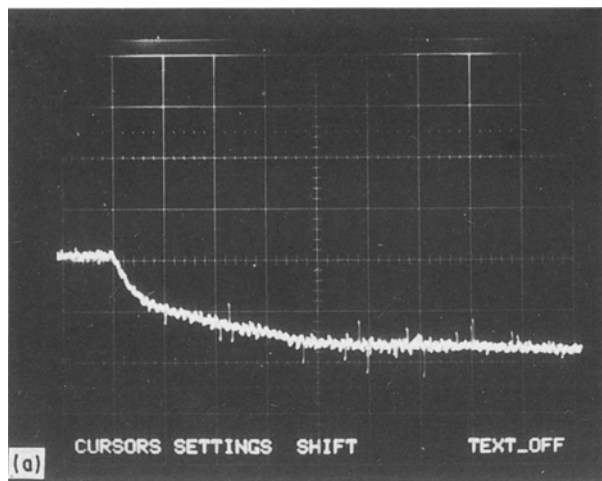
Figure 1 Experimentally obtained thermocouple voltage versus cooling time for alumina quenched into (a) water at $\Delta T = 350\,°C$ where each division on the x axis represents 6.25 ms and each division on the y axis represents 5 mV; (b) oil at $\Delta T = 480\,°C$ where each division on the x axis represents 200 ms and each division on the y axis represents 5 mV; and (c) liquid nitrogen at $\Delta T = 696\,°C$ where each division on the x axis represents 2 s and each division on the y axis represents 5 mV.

voltage. Using fourth-order polynomials, the derivative of the estimated temperature, $d\hat{T}/dt$, is given by

$$\frac{d\hat{T}}{dt} = \frac{1}{\Delta t}\left[\beta_2 + 2\beta_3\left(\frac{t}{\Delta t}\right) + 3\beta_4\left(\frac{t}{\Delta t}\right)^2 + 4\beta_5\left(\frac{t}{\Delta t}\right)^3\right] \quad (3)$$

The estimated heat transfer coefficient, $\hat{h}$, thus can be written as

$$\hat{h} = \frac{\rho C_p V}{A(T_\infty - \hat{T})}\left(\frac{d\hat{T}}{dt}\right) = \frac{mC_p}{A(T_\infty - \hat{T})}\left(\frac{d\hat{T}}{dt}\right) \quad (4)$$

where $m = \rho V =$ the specimen mass. The temperature-dependent terms in Equation 4 are thus C_p, the heat capacity, and A, the heated surface area of the specimen. The temperature dependence of A will be given by

$$A = A_0[1 + 2\alpha(T)\Delta T] \quad (5)$$

where $\alpha(T)$ is the coefficient of linear thermal expansion and A_0 is the surface area at some reference temperature. For alumina, the $2\alpha(T)\Delta T$ term will be very small with respect to unity over the temperature range included in this experiment. Thus to a good approximation, $A = A_0$ for this work and we therefore consider A to be temperature-independent. The heat capacity C_p, however, can vary significantly as a function of temperature. The available heat capacity data for polycrystalline alumina [5] was fitted via a least-squares technique to the polynomial form

$$C_p = c_1 + c_2 T + c_3 T^2 + c_4 T^3 \quad (6)$$

where C_p is the specific heat, T is the temperature (°C) and c_1, c_2, c_3 and c_4 are constants obtained from the linear regression analysis. The best-fit results for the constants c_1, c_2, c_3 and c_4 were $791.4\,J\,kg^{-1}\,°C^{-1}$, $1.407\,J\,kg^{-1}\,°C^{-2}$, $-1.634 \times 10^{-3}\,J\,kg^{-1}\,°C^{-3}$ and $7.029 \times 10^{-7}\,J\,kg^{-1}\,°C^{-4}$, respectively. The correlation coefficient for the C_p data fit to Equation 6 was 0.995 [1, 2].

4. Results and discussion

After the specimens were uniformly heated to a preselected temperature, they were quenched into a room-temperature water bath, into a room-temperature silicone oil bath, or into a liquid nitrogen bath. Using the polynomial coefficients β_i (Table I) obtained from the regression of the time versus temperature data (Equation 2), the heat transfer coefficients for each quenching medium were estimated using Equations 2–6.

As shown in this study's data (Figs 2–4) and in other researchers' data [6–11] (see also Figs 5–8 below), the surface heat transfer coefficient h varies significantly with the instantaneous temperature difference between the specimen surface and the fluid medium. In addition, when the quench medium begins to boil, the heat transfer is strongly affected by the nucleation of bubbles which increases the heat transfer. Gaseous film formation at high temperature differences suppresses the transfer of heat since the thermal conductivity of a gas is very low [9]. Except for recent work on water quenching by the present authors [1], the literature lacks direct measurements of the surface heat transfer coefficients of ceramics. In fact, there are only limited data available for the surface heat transfer coefficient for heat transfer at high temperatures into liquids other than water. For the silicone oil and liquid nitrogen quenching baths, we shall compare our

TABLE I Polynomial coefficients for measured temperature change versus time for polycrystalline alumina (Equation 2)

Quench medium	ΔT (°C)	β_1 (°C)	β_2 (°C)	β_3 (°C)	β_4 (°C)	β_5 (°C)	Data points taken	Correlation coefficient
Water (2.5 ms)[a]	250	258.60	− 18.88	1.09	− 0.03	3.24×10^{-4}	37	0.997
	300	309.34	− 21.90	1.33	− 0.04	3.91×10^{-4}	40	0.994
	350	365.74	− 28.33	1.76	− 0.05	5.53×10^{-4}	39	0.994
Oil[b] (20 ms)[a]	480	503.31	− 31.27	1.09	0.09	$- 4.19 \times 10^{-3}$	19	0.991
		284.14	− 3.31	0.06	− 0.001	7.3×10^{-6}	12	0.994
	510	535.10	− 33.24	0.81	0.11	$- 4.32 \times 10^{-3}$	17	0.996
		315.24	− 2.51	0.01	− 0.0001	7.2×10^{-7}	17	0.991
	560	586.05	− 35.91	0.36	0.19	$- 8.17 \times 10^{-3}$	20	0.991
		323.69	− 3.71	0.08	− 0.0075	1.2×10^{-5}	17	0.972
Liquid nitrogen (400 ms)[a]	696	491.85	− 22.86	0.95	− 0.036	5.21×10^{-4}	26	0.996
	551	351.69	− 20.58	1.29	− 0.069	1.30×10^{-3}	20	0.997
	305	107.2	− 8.83	0.34	− 0.037	1.49×10^{-4}	10	0.998

[a] The time specified in parenthesis represents the time step Δt used in the regression analysis (Equation 2).
[b] The entire data set for the silicone oil quench is not well described by a single fourth-order polynomial (Equation 2, with $p = 5$). Thus, we separated the data into two sets. This separation reflects the change in the slope of h versus temperature (Fig. 3), which may in turn represent a change in the heat transfer mechanism (see section 4, also Ozisik [12] and Holman [13].

alumina results with the heat transfer coefficients measured for metals.

The heat transfer coefficient values obtained for alumina in this study are roughly similar to both the heat transfer coefficient values calculated for water-quenched alumina by Becher and co-workers [6, 7] (Fig. 5) and to the heat transfer coefficient values inferred from experimental critical temperature differences for water-and silicone oil-quenched alumina by Singh *et al.* [8] (Fig. 6).
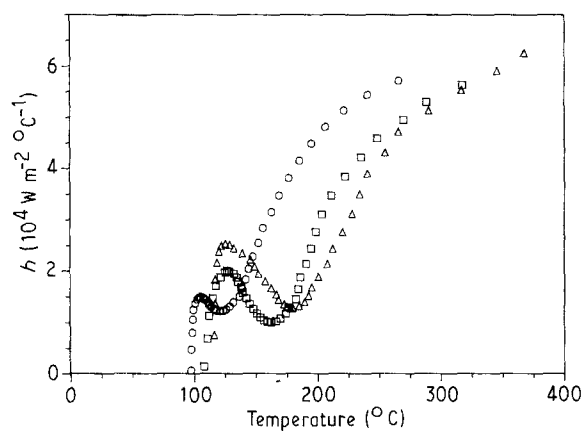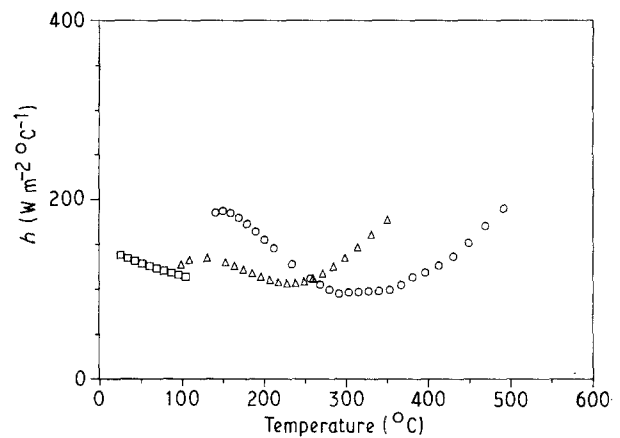

Figure 2 Estimated surface heat transfer coefficient versus temperature calculated from the experimentally obtained thermocouple voltage versus cooling time for water-quenched alumina: (○) $\Delta T = 250\,°C$, (□) $\Delta T = 300\,°C$, (△) $\Delta T = 350\,°C$ (after [1]).
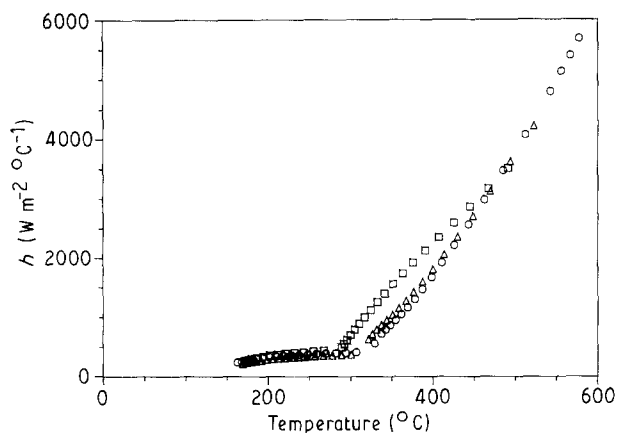

Figure 4 Estimated surface heat transfer coefficient versus temperature calculated from the experimentally obtained thermocouple voltage versus cooling time for liquid nitrogen-quenched alumina: (□) $\Delta T = 305\,°C$, (△) $\Delta T = 551\,°C$, (○) $\Delta T = 696\,°C$.

Figure 3 Estimated surface heat transfer coefficient versus temperature calculated from the experimentally obtained thermocouple voltage versus cooling time for silicone oil-quenched alumina: (□) $\Delta T = 480\,°C$, (△) $\Delta T = 510\,°C$, (○) $\Delta T = 560\,°C$.
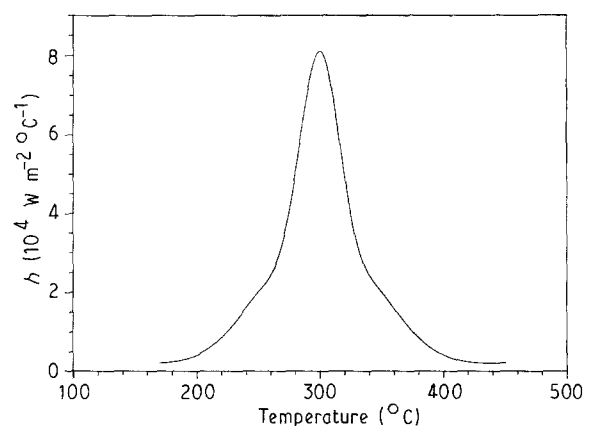

Figure 5 Surface heat transfer coefficient of alumina for water quench inferred from the experimental ΔT_c values (after Becher [6]).
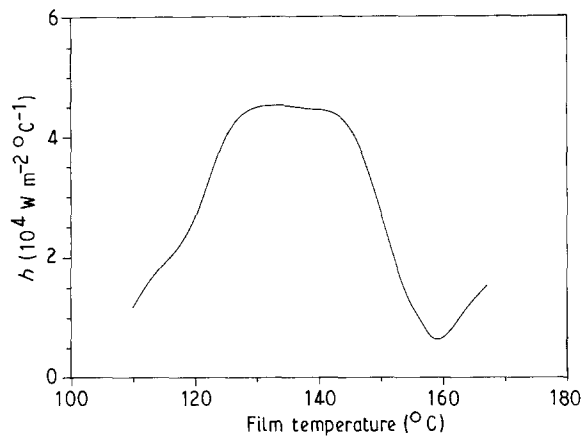
Figure 6 Surface heat transfer coefficient of alumina for water quench inferred from the experimental ΔT_c values (after Singh *et al.* [8]).

Figure 8 Surface heat transfer coefficient of an Inconel plate quenched into liquid nitrogen for liquid (after Sauer and Ragsdell [14]).

Stolz *et al.* [10] heated a 2.54 cm radius silver sphere to 870 °C and quenched it into a 43 °C oil bath. Stolz *et al.* measured the interior temperature (0.254 cm from the sphere's surface) versus time and found the surface heat flux versus time. The heat transfer coefficient for Stolz's [10] silver sphere quenched into oil was evaluated as a function of temperature up to about 870 °C (Fig. 7). The maximum heat transfer coefficient (about 11000 $W\,m^{-2}\,°C^{-1}$) occurred for a surface temperature range of 520–530 °C. The heat transfer coefficient increased by well over an order of magnitude between about 150 and 560 °C (Fig. 7). Hachisu *et al.* [11] used a nickel–silver thermocouple to obtain similar values for the temperature dependence of heat transfer coefficient for a steel cylinder quenched into oil. The heat transfer coefficient was evaluated from an unsteady-state heat transfer analysis of the resulting cooling curve. Hachisu *et al.* found a maximum heat transfer coefficient of 3000 $W\,m^{-2}\,°C^{-1}$ in the surface temperature range of 500–600 °C (Fig. 7).

In this study, the heat transfer coefficient of alumina quenched into silicone oil increased as the surface
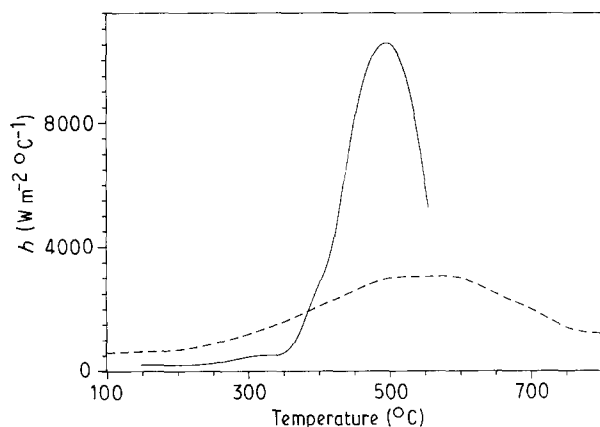

Figure 7 Surface heat transfer coefficient of (——) a silver sphere for an oil quench (after Stolz *et al.* [10]) and (– – –) a steel cylinder for an oil quench, after Hachisu *et al.* [11]).

temperature increased up to 600 °C (Fig. 3). The maximum heat transfer coefficient in this study was about 6000 $W\,m^{-2}\,°C^{-1}$, which was lower than the value of 11000 $W\,m^{-2}\,°C^{-1}$ found by Stolz *et al.* [10] and higher than the value of 3000 $W\,m^{-2}\,°C^{-1}$ found by Hachisu *et al.* [11].*

A change in the slope of the *h* versus temperature curve can indicate a change in the heat transfer mechanism [12, 13]. For our data (Fig. 3), the change in the slope of the *h* versus temperature curve may indicate that upon quenching into the silicone oil bath, the heat transfer mechanism for the alumina specimens changes from convection accompanied by boiling nucleation to a pure convection mechanism at roughly 300 to 350 °C. This transition point was observed in the data of Stolz *et al.* at about 380 °C and in those of Hachisu *et al.* at about 200 °C (Fig. 7).

For quenching into a liquid nitrogen bath, the heat transfer phenomenon between liquid nitrogen and a solid is complicated because when a hot specimen reaches the liquid nitrogen bath, the surface is immediately shielded by a film of nitrogen gas. Due to the film formation, the surface heat transfer rate becomes very small (Fig. 4).

In a study of nitrogen film boiling from a flat surface, Sauer and Ragsdell [14] measured the heat flux for an Inconel 600 plate as a function of the plate temperature for the temperature range from about 40 °C up to 593 °C. Six Chromel–Alumel thermocouples were attached to the Inconel plate. The plate and a heater assembly were placed into a liquid nitrogen-filled Dewar flask. Power to the heater assembly was increased slowly until film boiling was initiated. Data recorded after equilibrium was reached indicated that the heat transfer coefficient was nearly constant at about 290 $W\,m^{-2}\,°C^{-1}$ up to 590 °C (Fig. 8) [14].

In this study alumina specimens were dipped into a liquid nitrogen bath and the transient surface temperature was measured. For initial specimen temperatures between 30 and 500 °C, the value of *h*

*Stolz *et al.* [10] and Hachisu *et al.* [11] did not specify the chemical composition of their quenching oils or any of their quenching oils' properties. The heat transfer coefficient depends on fluid properties such as the fluid's coefficient of thermal expansion, density, specific heat, and thermal conductivity as well as the fluid's viscosity [12, 13].
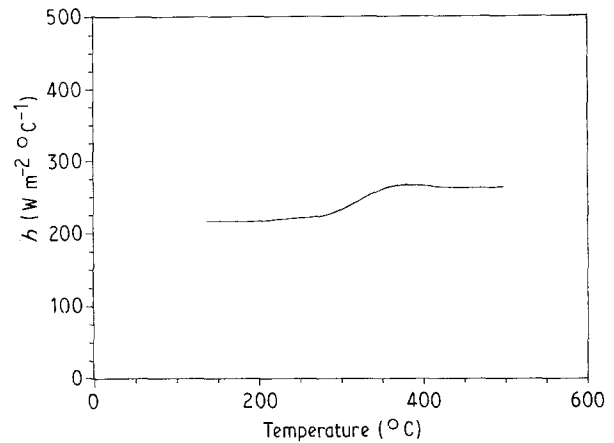
TABLE II Maximum heat transfer coefficient for various quench media

Quench medium	Maximum value of h (W m^{-2} °C^{-1})	Temperature (°C)[a]	Material	Specimen geometry	Reference
Water	56 000	220	Steel	Cylinder	[3]
	63 000	370	Alumina	Bar	This study
	53 000	410	TiB$_2$	Bar	[1]
	200 000	370	Macor	Bar	[1]
Oil	11 000	530	Silver	Sphere	[10]
	3000	550	Steel	Cylinder	[11]
	6000	600	Alumina	Bar	This study
Liquid nitrogen	290	550–660	Inconel	Plate	[14]
	200	500	Alumina	Bar	This study

[a] The temperature indicated corresponds to the temperature at which the maximum h value was measured.

determined in this study was between 100 and 200 W m^{-2} °C^{-1}, which agrees reasonably well with the 290 W m^{-2} °C^{-1} value obtained for Inconel by Sauer and Ragsdell [14]. The maximum heat transfer coefficient for the oil bath was an order of magnitude lower than that for the water bath and an order of magnitude higher than that for the liquid nitrogen bath (Table II).

5. Summary and conclusions

Values of the surface heat transfer coefficient h were measured for alumina quenched into water, into silicone oil, and into liquid nitrogen. The measurements show that the maximum h for the silicone oil quench (about 6000 W m^{-2} °C^{-1}) is an order of magnitude higher than h for the liquid nitrogen quench (about 200 W m^{-2} °C^{-1}) and an order of magnitude lower than h for the water quench (about 60 000 W m^{-2} °C^{-1}).

For both the water and the silicone oil quenches, h changed rapidly as a function of the instantaneous surface temperature of the alumina specimens. In contrast, the liquid nitrogen quench, with its rapid formation of a gaseous film, had a relatively low and constant value of h. Except for one recent study by the present authors that includes direct measurements of h for water quenching of selected ceramics [1], the literature lacks direct h measurements for alumina (or other ceramics) quenched into water, into silicone oil, or into liquid nitrogen. However, this study's values of h for alumina quenched into water are roughly comparable with the water-quench h values for alumina that are estimated from free convection calculations [6, 7] or from fracture mechanics-based estimates [8]. For silicone oil quenching, this study's h values are comparable to those estimated from critical quench difference calculations [8]. The literature does include direct h measurements for metals and alloys quenched

into liquid nitrogen baths [14] and into oil baths [10, 11] that show h magnitudes and temperature trends that are broadly comparable to the h measurements for alumina quenched into liquid nitrogen and silicone oil.

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